Calculation of (P,T)-odd electric dipole moments for the diamagnetic atoms ^{129}Xe, ^{171}Yb, ^{199}Hg, ^{211}Rn, and ^{225}Ra

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Electric dipole moments of diamagnetic atoms of experimental interest are calculated using the relativistic Hartree-Fock and random-phase approximation methods, the many-body perturbation theory, and the configuration-interaction technique. We consider (P,T)-odd interactions, which give rise to atomic electric dipole moment in the second order of the perturbation theory. These include nuclear Schiff moment, (P,T)-odd electron-nucleon interaction, and electron electric dipole moment. Interpretation of an experimental constraint of a permanent electric dipole moment of ^{199}Hg [W. C. Griffith, M. D. Swallows, T. H. Loftus, M. V. Romalis, B. R. Heckel, and E. N. Fortson, Phys. Rev. Lett. **102**, 101601 (2009)] is discussed.

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I. INTRODUCTION

The search for a permanent electric dipole moment (EDM) of particles, violating both parity and time-reversal invariance, has a long history (see, e.g., [1]). The standard model predicts tiny EDMs, which cannot be detected at the present level of experimental accuracy. However, different extensions of the standard model (such as, e.g., supersymmetry) predict much larger EDMs of the particles that, in principle, could be found using the modern experimental technique. A reveal of such EDMs would unambiguously lead to a contradiction with the standard model.

According to [2,3] previous limits on EDM produced stringent constraints on electroweak bariogenesis and models of *CP* violation, but fall short of ruling out the simplest generic extensions of the standard model. It was stated that the next generation of EDM experiments should be sufficiently sensitive to provide a conclusive test.

A very significant step ahead in this direction has been done in a recent work [4]. The authors reported result obtained for a permanent EDM of ^{199}Hg to be $d(^{199}\mathrm{Hg}) = (0.49 \pm 1.29_{\mathrm{stat}} \pm 0.76_{\mathrm{syst}}) \times 10^{-29} e$ cm. Although the EDM is still consistent with zero, its limit $|d(^{199}\mathrm{Hg})| < 3.1 \times 10^{-29}|e|$ cm is an improvement of the previous Hg limit by a factor of 7. Motivated by this result and implying future experimental progress, we have performed calculations of different contributions to the atomic EDMs of ^{129}Xe, ^{171}Yb, ^{199}Hg, ^{211}Rn, and ^{225}Ra. For these atoms the experiments searching for EDMs are underway.

The paper is organized as follows. In Sec. II we discuss different types of (P,T)-odd interactions that can lead to an appearance of a permanent atomic EDM. In Sec. III we describe the methods of calculations of the EDMs. We start our calculations from the relativistic Hartree-Fock method. Than we include many-body corrections using two different methods. First, we apply a simple random-phase approximation (RPA) for the closed-shell atoms. Second, we apply the configuration interaction (CI) combined with the many-body perturbation theory (MBPT) [12] approach to valence electrons while we use the RPA approach for the core. Section IV is devoted to an analysis and discussion of the results. We present the results obtained for different contributions to the atomic EDMs and compare them with other available data. In Sec. V we discuss the neutron and the proton contributions to the total nuclear spin using the spherical shell model of a nucleus. Section VI contains concluding remarks and two final tables where the recommended values of the contributions to the EDMs of ^{129}Xe, ^{171}Yb, ^{199}Hg, ^{211}Rn, and ^{225}Ra are gathered and the limits on *CP*-violating parameters based on an experimental limit for ^{199}Hg are presented.

II. GENERAL FORMALISM

Our goal is to find the atomic EDM d_{at} defined as $\mathbf{d}_{\mathrm{at}} \equiv d_{\mathrm{at}}(\mathbf{F}/F)$, where $\mathbf{F}=\mathbf{J}+\mathbf{I}$ with $\mathbf{J}$ being total angular momentum. In this work we deal with the atoms with closed shells in their ground states. In this case $\mathbf{J}=0$, $\mathbf{F}=\mathbf{I}$, and $\mathbf{d}_{\mathrm{at}} = d_{\mathrm{at}}(\mathbf{I}/I)$.

We consider several types of (P,T)-odd interactions between particles leading to an appearance of an atomic EDM. We restrict ourselves to contributions to the EDM which occur in the second order of the perturbation theory. The EDM induced in an atomic state $|0\rangle$ due to an admixture of opposite-parity states that appears in the second order of the perturbation theory can be written as

$$\mathbf{d}_{\mathrm{at}} = 2\sum_K \frac{\langle 0|\mathbf{D}|K\rangle\langle K|H|0\rangle}{E_0 - E_K}, \tag{1}$$

where $\mathbf{D}=-|e|\mathbf{r}$ is the electric dipole operator, e is the electron charge, and E_i are the energies of the states.

We will consider below (1) the tensor-pseudotensor (P,T)-odd electron-nucleon $(e\text{-}N)$ interaction, (2) the pseudoscalar-scalar (P,T)-odd $e\text{-}N$ interaction, (3) the nuclear Schiff moment, and (4) the interaction of the electron EDM with the internal nuclear magnetic field of the atom.

It is worth noting that the operators describing all these interactions have certain similar features. All of them (1) are proportional to the nuclear spin $\mathbf{I}$ and (2) have strong singularity. The similar nature of these operators leads to a suggestion that the calculations depend very little on their specific form. It allows us to expect that the relative

contributions of different many-body parts of calculations will remain approximately the same for all operators. As we will show below this suggestion is fully justified.

A. Electron-nucleon (P,T)-odd interactions

We start our consideration with brief reminder of the main features of the e-N (P,T)-odd interaction leading to appearance of atomic electric dipole moments in the second order of the perturbation theory. A detailed description can be found elsewhere [1,5]. This interaction has the following form (see, e.g., [5]):

$$H = \frac{G}{\sqrt{2}} \sum_N [C_T^N \bar{N} i\gamma_5 \sigma_{\mu\nu} N \bar{e} \gamma_5 \sigma_{\mu\nu} e + C_P^N \bar{N} i\gamma_5 N \bar{e} e]. \quad (2)$$

Here, C_T^N and C_P^N are dimensionless coupling constant characterizing tensor-pseudotensor and pseudoscalar-scalar (P,T)-odd electron-nucleon interactions for the nucleon N, $\sigma_{\mu\nu}=(\gamma_\mu\gamma_\nu-\gamma_\nu\gamma_\mu)/2$, and γ_5 and $\boldsymbol{\gamma}$ are the Dirac matrices $\gamma_5=\left(\begin{smallmatrix}0 & -1\\ -1 & 0\end{smallmatrix}\right)$ and $\boldsymbol{\gamma}=\left(\begin{smallmatrix}0 & \boldsymbol{\sigma}\\ -\boldsymbol{\sigma} & 0\end{smallmatrix}\right)$.

In accordance with Eq. (2) we can represent the Hamiltonian H as $H \equiv H_T + H_P$, where in the coordinate representation (atomic units $\hbar = m_e = |e| = 1$ are used throughout)

$$H_T = i\sqrt{2} G C_T \boldsymbol{\gamma} \langle \boldsymbol{\sigma}_N \rangle \rho(r), \quad (3)$$

$$H_P = -\frac{G}{\sqrt{2}} \frac{1}{2m_p c} C_P \gamma_0 \nabla \rho(r) \langle \boldsymbol{\sigma}_N \rangle. \quad (4)$$

Here, G is the Fermi constant, c is the speed of light (in atomic units $c = 1/\alpha \approx 137$), and m_p is the nucleon mass.

We denote

$$C_T \langle \boldsymbol{\sigma}_N \rangle \equiv \left\langle C_T^p \sum_p \boldsymbol{\sigma}_p + C_T^n \sum_n \boldsymbol{\sigma}_n \right\rangle,$$

$$C_P \langle \boldsymbol{\sigma}_N \rangle \equiv \left\langle C_P^p \sum_p \boldsymbol{\sigma}_p + C_P^n \sum_n \boldsymbol{\sigma}_n \right\rangle,$$

where $\langle \cdots \rangle$ means averaging over the nuclear state with the nuclear spin $\boldsymbol{I}$.

In Eq. (4) we keep only the term in the lowest nonvanishing approximation in m_p^{-1}. Note that in this equation the operator ∇ acts only to $\rho(r)$, where $\rho(r)$ is the nuclear density distribution.

Since we are dealing with very singular operators the model of the nuclear density distribution can be important. To check this point we have carried out calculations for two models. In one of them the nucleus was treated as a charged sphere with the radius R, i.e.,

$$\rho(r) = \frac{3}{4\pi R^3} \theta(R - r). \quad (5)$$

In other model it was used the Fermi distribution

$$\rho(r) = \frac{\rho_0}{1 + \exp\dfrac{r - R}{a}}, \quad (6)$$

where ρ_0 is the normalization parameter determined by $\int \rho dV = Z$. We have found that the results obtained for each of these models were numerically very close to each other.

Similar to the expression for H it is convenient to represent $\mathbf{d}_{\rm at}$ as a sum of two terms $\mathbf{d}_{\rm at} = \mathbf{d}_{\rm at}^T + \mathbf{d}_{\rm at}^P$, where $\mathbf{d}_{\rm at}^T$ and $\mathbf{d}_{\rm at}^P$ correspond to the operators H_T and H_P given by Eqs. (3) and (4). Explicit expressions for $\mathbf{d}_{\rm at}^T$ and $\mathbf{d}_{\rm at}^P$ can be derived from Eq. (1) by replacing the operator H to H_T and H_P, correspondingly.

It is convenient to determine the quantities $d_{\rm at}^T$ and $d_{\rm at}^P$ as follows: $\mathbf{d}_{\rm at}^{T,P} = d_{\rm at}^{T,P} \langle \boldsymbol{\sigma}_N \rangle \sim d_{\rm at}^{T,P} \mathbf{I}/I$. The coefficient of proportionality in this expression depends on a model of the nucleus. An accurate treatment of the nuclear structure is beyond the topic of this work. For a spherical shell model of the nucleus this coefficient can be easily found for different atoms. We will discuss this problem in more detail in Sec. V.

B. Nuclear Schiff moment

The Schiff moment is a nuclear moment violating both parity and time-reversal invariance. It is caused by (P,T)-odd nuclear forces and it takes into account screening of external electric field by atomic electrons. We use the form of the Hamiltonian for the interaction of atomic electrons with the nuclear Schiff moment suggested in Ref. [6]

$$H_{\rm SM} = -\frac{3\mathbf{S}\mathbf{r}}{B} \rho(r), \quad (7)$$

where $B \equiv \int \rho(r) r^4 dr$ and $\mathbf{S}$ is the Schiff moment vector defined as $\mathbf{S} = S(\mathbf{I}/I)$ with S being the coupling constant.

Its contribution to the EDM of the closed-shell atoms has been considered in detail in [7,8]. As it follows from [7] the results obtained in the frame of multiparticle approach combining the CI with the MBPT agreed (within 10%) with the results obtained by the RPA method. Here, we perform similar calculations mostly for consistency test and for completeness.

C. Electron EDM

An interaction of the electron EDM d_e with the electromagnetic field strength $F_{\mu\nu}$ can be written in the relativistically covariant form as

$$H_e = \frac{d_e}{2} \bar{\psi} \gamma_5 \sigma_{\mu\nu} \psi F_{\mu\nu}. \quad (8)$$

Here, $\bar{\psi} = \psi^\dagger \gamma_0$ and ψ is determined in the Appendix.

Again we will consider here only the effect appearing in the second order of the perturbation theory. It is characterized by an interaction of the electron EDM with the magnetic field $\mathbf{B}$ created by the nuclear magnetic moment. The operator of this interaction (H_B) can be written as

$$H_B = -i d_e \boldsymbol{\gamma} \mathbf{B}. \quad (9)$$

The magnetic field $\mathbf{B}$ can be represented by

$$\mathbf{B} = \boldsymbol{\nabla} \times \frac{(\mathbf{M} \times \mathbf{r})}{r_>^3} = \frac{3(\mathbf{M} \cdot \mathbf{n})\mathbf{n} - \mathbf{M}}{r^3} \theta(r - R) + \frac{2\mathbf{M}}{R^3} \theta(R - r), \quad (10)$$

where

$$\theta(x) = \begin{cases} 1, & x \geq 0 \\ 0, & x < 0 \end{cases}$$

and $\mathbf{n} \equiv \mathbf{r}/r$. The contribution to the atomic EDM can be written as $\mathbf{d}_{\rm at}^B = d_{\rm at}^B(\mathbf{I}/I)$ and found from Eq. (1) by replacing $H \to H_B$.

III. METHODS OF CALCULATION

A. RPA for the closed shells

Here, we describe a simple method suitable for calculations of atomic EDM for atoms with closed shell. On the first stage we solve Dirac-Hartree-Fock (DHF) equations in the V^N approximation (i.e., including all electrons forming the ground state of the atom in a self-consistency procedure)

$$\hat{H}_0 \psi_c = \varepsilon_c \psi_c. \tag{11}$$

Here, H_0 is the relativistic Hartree-Fock Hamiltonian and ψ_c and ε_c are single-electron wave functions and energies.

At the next step we construct virtual orbitals. Different techniques can be used for this procedure. One approach is to multiply the previous orbital of the same partial wave to a smooth function of r with subsequent orthogonalization of this orbital to all the rest orbitals. This method was described in detail in Refs. [9,10].

Another method is to construct a basis set using the B-spline technique developed at the University of Notre Dame [11]. We use 60 B splines of order 9 in a cavity of radius $R_{\rm max} = 30 a_B$, where a_b is the Bohr radius. This relatively large number of B splines is needed due to singularity of the (P,T)-odd operators. This requires a very detailed description of the wave functions in the vicinity of the nucleus.

Further, we consider an atom in external field and solve the RPA equations (self-consistent DHF in an external field)

$$(\hat{H}_0 - \varepsilon_c)\delta\psi_c = -(\hat{F} + \delta V_F^N)\psi_c, \tag{12}$$

where $\hat{F}$ is the operator of external field and δV_F^N is the correction to the self-consistent potential due to the effect of external field. The index c numerates single-electron functions (ψ_c) of the closed-shell core. The RPA equations (12) are solved self-consistently for all states in the core for all external fields involved in the problem.

B. CI+MBPT

A more sophisticated and accurate way to calculate atomic EDMs is to use the configuration-interaction technique for valence electrons while still using the RPA approach for the core. This would allow us to check the accuracy of the RPA calculations. It is especially important for the atoms having two external s electrons: Yb, Hg, and Ra. The effect of external electrons on different properties of these atoms is large, and an accurate treatment of the interaction between them is needed.

We consider Yb, Hg, and Ra as atoms with two valence electrons above closed-shell cores $[1s,\ldots,4f^{14}]$, $[1s,\ldots,5d^{10}]$, and $[1s,\ldots,6p^6]$, respectively. In this paper we follow approach suggested in [12] which combines the many-body perturbation theory with the configuration-interaction method. We will refer to it as the CI+MBPT formalism. The MBPT is used to include excitations from the core into the effective Hamiltonian for valence electrons. After that, the multiparticle relativistic equation for valence electrons is solved within the CI framework to find the wave functions and the low-lying energy levels.

In the CI+MBPT method, the energies and the wave functions are determined from the time-independent equation

$$H_{\rm eff}(E_n)\Phi_n = E_n \Phi_n, \tag{13}$$

where the effective Hamiltonian is defined as

$$H_{\rm eff}(E) = H_{\rm FC} + \Sigma(E). \tag{14}$$

Here, $H_{\rm FC}$ is the Hamiltonian in the frozen core approximation and Σ is the energy-dependent correction, which takes into account virtual core excitations. The operator Σ completely accounts for the second-order perturbation theory over residual Coulomb interaction.

Since we are interested in calculating the atomic EDMs, we need to construct the corresponding effective operators for valence electrons [13–15]. To do that, we can extend the concept of the effective Hamiltonian $H_{\rm eff}$ to other operators such as the effective dressed electric-dipole operator $D_{\rm eff}$ and the (P,T)-odd operators. These operators account for the core-valence correlations. As in pure RPA approach of Sec. III A, we solve the RPA equations summing a certain sequence of many-body diagrams to all orders of MBPT [13,16,17]. Since requirements to the accuracy of calculations are not very high, we disregard in this consideration small corrections like normalization and structural radiation.

We perform the calculations in the CI+MBPT method in V^{N-1} and V^{N-2} potentials. The former is a bit more "natural" for the lowest-lying odd-parity states of the considered atoms such as $nsnp\ ^{3,1}P_1^o$ ($n=6$ for Yb and Hg and $n=7$ for Ra), because $6p_{1/2,3/2}$ (or $7p_{1/2,3/2}$) orbitals are constructed at the stage of solving DHF equations for the configuration $[\text{core}]nsnp$. The latter is somewhat simpler (e.g., due to an absence of the subtraction diagrams). We have checked that the final results in both potentials are in good agreement with each other. For this reason when we discuss results, we do not distinguish between these potentials.

Two different basis sets described in Sec. III A were used in the V^{N-1} and V^{N-2} approximations. First set [9,10] was used in the V^{N-1} and B-spline set [11] was used in the V^{N-2} calculations. At the CI stage of the V^{N-1} calculations the one-electron basis set for Yb included $1s$-$13s$, $2p$-$13p$, $3d$-$12d$, $4f$-$11f$, and $5g$-$7g$ orbitals, where the core and $5d$ and $6p$ orbitals are Dirac-Hartree-Fock ones and all the rest are the virtual orbitals. For Hg and Ra the basis sets were insignificantly larger. In all cases the basis sets were numerically complete and the full CI was made for two valence electrons. At the stage of the MBPT calculations a more extended basis sets, including more basis functions, were used. For instance, for Yb it included $1s$-$26s$, $2p$-$26p$, $3d$-$25d$, $4f$-$17f$, and $5g$-$12g$ orbitals.

All B splines up to $l_{\rm max}=5$ were used to calculate Σ in the V^{N-2} approximation. 18 lowest basis states above the core in

TABLE I. The isotopes of the atoms considered in this work. μ are the magnetic moments expressed in nuclear magnetons.

	Z	A	I	μ
Xe	54	129	1/2	−0.7778
Yb	70	171	1/2	0.4119
Hg	80	199	1/2	0.5059
Rn	86	211	1/2	0.60
Ra	88	225	1/2	−0.734

each partial wave up to $l_{\max}=3$ were used on the CI stage of the V^{N-2} calculations.

IV. RESULTS AND DISCUSSION

A. RPA for the closed shells

In the frame of the RPA method discussed in Sec. III we have performed the calculations of different contributions to the EDMs of the diamagnetic atoms presented in Table I. For these atoms the experiments searching for the EDMs are planned or are underway.

We can rewrite Eq. (1) as follows:

$$\mathbf{d}_{\rm at}=2\sum_{a,k}\frac{\langle k|\mathbf{r}|a\rangle\langle k|H|a\rangle}{\varepsilon_a-\varepsilon_k}, \tag{15}$$

where the summation is over the quantum numbers of the one-electron core states "a" and excited states "k" and ε_i are the one-electron energies.

1. (P,T)-odd e-N interaction

We start the discussion from tensor-pseudotensor and pseudoscalar-scalar e-N (P,T)-odd interactions. Using the Wigner-Eckart theorem and going over to the reduced matrix elements (ME), we obtain for the contributions $d_{\rm at}^T$ and $d_{\rm at}^P$ to atomic EDM

$$d_{\rm at}^T=\frac{2\sqrt{2}G}{3}C_T\sum_{a,k}\frac{\langle k||r||a\rangle\langle k||i\gamma\rho(r)||a\rangle}{\varepsilon_k-\varepsilon_a}, \tag{16}$$

$$d_{\rm at}^P=-\frac{GC_P}{3\sqrt{2}m_pc}\sum_{a,k}\frac{\langle k||r||a\rangle\langle k||n(d\rho/dr)\gamma_0||a\rangle}{\varepsilon_k-\varepsilon_a}. \tag{17}$$

The explicit expressions for the reduced MEs of the (P,T)-odd operators in Eqs. (16) and (17) are given in the Appendix.

In Table II we present the values of $d_{\rm at}^T$ obtained for all isotopes listed in Table I in pure DHF approximation and including RPA corrections. Note that the RPA corrections must be included only for one operator in Eq. (16) or Eq. (17) [18]. In other words, when we include the RPA corrections for the electric dipole operator r, we must not include them for the (P,T)-odd operator and vice versa. Certainly both approaches should lead to the same result. It allows us to test the consistency of the calculations.

Our results for Xe are in good agreement with other data. Our RPA value for ^{199}Hg is in excellent agreement with similar calculations by Maartensson-Pendrill [18] while the result obtained in Ref. [19] is somewhat larger. There is also a reasonable agreement between the result found in this work and the estimate obtained by Sushkov *et al.* from the analytically derived formula [20]. To the best of our knowledge there are no other data for Yb, Ra, and Rn to compare with.

TABLE II. The values of $\mathbf{d}_{\rm at}^T$ in units $(10^{-20}C_T\langle\boldsymbol{\sigma}_N\rangle|e|$ cm) obtained in DHF and RPA approximations are presented. The results are compared with other data.

	^{129}Xe	^{171}Yb	^{199}Hg	^{211}Rn	^{225}Ra
This work (DHF)	0.45	−0.70	−2.4	4.6	−3.5
Ref. [18] (DHF)	0.41		−2.0		
Ref. [21] (DHF)	0.41				
Ref. [20]	0.6		−3.9		
This work (RPA)	0.57	−3.4	−5.9	5.6	−17
Ref. [18] (RPA)	0.52		−6.0		
Ref. [19] (RPA)			−6.75		

As is seen from Table II, the inclusion of the RPA corrections leads to increasing atomic EDM. For the noble gases (Xe and Rn) the RPA corrections contribute at the level of 30%, while for the atomic Hg, Yb, and Ra, which have two s electrons above closed shells, the RPA corrections are much larger. In fact, they increase the EDM 2.5 times for Hg and five times for Yb and Ra as compared to the DHF results. The reason for this increase is that the two s electrons are loosely bound and can be easily excited. As a result, an account for the higher orders of the perturbation theory (like the RPA corrections) leads to significant change in the "bare" results obtained in the DHF approximation.

The results obtained for $\mathbf{d}_{\rm at}^P$ are listed in Table III. In Ref. [22] Flambaum and Khriplovich suggested a method to establish the correspondence between contributions of the tensor-pseudotensor and pseudoscalar-scalar (P,T)-odd operators using the expressions for the reduced MEs of these operators (see the Appendix).

This correspondence can be obtained using the properties of the wave functions f_s, g_s, $f_{p_{1/2}}$, and $g_{p_{1/2}}$ in the vicinity of the nucleus (see, e.g., [6]). For instance,

$$f_{p_{1/2}}(r)\approx g_{p_{1/2}}(R)\tfrac{1}{2}Z\alpha x\left(1-\tfrac{1}{5}x^2\right), \tag{18}$$

where $x\equiv r/R$.

TABLE III. The values of $\mathbf{d}_{\rm at}^P$ in units $(10^{-23}C_P\langle\boldsymbol{\sigma}_N\rangle|e|$ cm) obtained in DHF and RPA approximations are presented. The results are compared with other data.

	^{129}Xe	^{171}Yb	^{199}Hg	^{211}Rn	^{225}Ra
This work (DHF)	1.3	−2.4	−8.7	17.3	−13.0
This work (RPA)	1.6	−11.5	−21.3	21.0	−63.7
Rescaling (RPA)[a]	1.6	−11.3	−21.3	21.3	−64.7

[a]These numbers are obtained using the correspondence $C_P\leftrightarrow C_T$ given by Eq. (20) as explained in the text.

TABLE IV. The values of $d_{\rm at}^{SM}$ in units $\{10^{-17}[S/(|e|{\rm fm}^3)]|e|\ {\rm cm}\}$ obtained in DHF and RPA approximations and in the frame of the coupled-cluster single-double (CCSD) method [23] are presented. The results are compared with other data.

	^{129}Xe	^{171}Yb	^{199}Hg	^{211}Rn	^{225}Ra
This work (DHF)	0.29	−0.42	−1.2	2.5	−1.8
This work (RPA)	0.38	−1.9	−3.0	3.3	−8.3
Ref. [7] (RPA)	0.38		−2.8	3.3	−8.5
Ref. [8] (RPA)		−1.9			
Ref. [23] (CCSD)			−5.07		

At $r=R$ we obtain

$$f_{p_{1/2}}(r) \approx g_{p_{1/2}}(R)\tfrac{1}{2}Z\alpha \times 0.8 \tag{19}$$

and after some transformations we find the correspondence

$$C_P \leftrightarrow \frac{5m_pR}{Z\alpha}C_T \approx 3.8\times 10^3\frac{A^{1/3}}{Z}C_T. \tag{20}$$

The final coefficient connecting C_P and C_T in Eq. (20) differs by a factor of 6/5 from that obtained in [22].

Given the results obtained in this work in the RPA approximation for $d_{\rm at}^T$ and using Eq. (20), we can find the values of $d_{\rm at}^P$. These values are listed in Table III in the entry "rescaling (RPA)." As is seen from the table there is an excellent agreement between the calculated and the rescaled values. It is worth noting that Eq. (20) turns out to be insensitive to Z. For the comparably light Xe and for heavy Ra the agreement of the numerical results and the results obtained with use of Eq. (20) is equally good. It means that Eq. (20) works well for atoms with different Z.

2. Schiff moment

The contribution to the atomic EDM due to the Schiff moment H^{SM} is naturally to determine as $\mathbf{d}_{\rm at}^{SM}=d_{\rm at}^{SM}(\mathbf{I}/I)$, where $d_{\rm at}^{SM}$ is given by

$$d_{\rm at}^{SM} = -2\frac{S}{B}\sum_{a,k}\frac{\langle k||r||a\rangle\langle k||r\rho(r)||a\rangle}{\varepsilon_k-\varepsilon_a}. \tag{21}$$

In explicit form the ME of the operator $[r\rho(r)]$ is given in the Appendix.

The results for the Schiff moment contribution to the atomic EDMs are presented in Table IV. There is a very good agreement with previous calculations [7,8]. Few-percent difference for Hg and Ra is within the accuracy of the calculations for these atoms.

A reason of the discrepancy with the result obtained in Ref. [23] is unclear for us. The authors of this work state that a possible reason of the difference between their result and those obtained in Ref. [7] is due to electron correlations which drastically change the final result. However, according to our calculations, the correlations included in the CI +MBPT approach change the results insignificantly, on the level of 10–15 % as compared to the RPA calculations. The same conclusion was made in Ref. [7]. Below we will return to this problem.

TABLE V. The values of $d_{\rm at}^B$ in units $(d_e\times 10^{-4})$ obtained in DHF and RPA approximations are presented. The results are compared with other data.

	^{129}Xe	^{171}Yb	^{199}Hg	^{211}Rn	^{225}Ra
This work (DHF)	0.85	1.0	4.9	−11	−11
Ref. [24] (DHF)	0.86		5.1		
This work (RPA)	1.0	5.1	12.5	−13	−55
Ref. [24] (RPA)	1.05		13		

3. Electron EDM

Usually in experiments searching for atomic EDMs atoms are placed in an external electric field $\mathbf{E}_{\rm ext}$. It leads to appearance of the interaction $\mathbf{d}_{\rm at}\mathbf{E}_{\rm ext}=-d_e\gamma_5\boldsymbol{\gamma}\mathbf{E}_{\rm ext}$. The operator $(\gamma_5\boldsymbol{\gamma})$ is P even and T odd. If the hyperfine interaction $H_{\rm hf}$ is accounted for, a contribution to the atomic EDM caused by this operator appears already in the second order of the perturbation theory and looks as follows:

$$2\sum_K \frac{\langle 0|H_{\rm hf}|K\rangle\langle K|d_e\gamma_5\boldsymbol{\gamma}|0\rangle}{E_0-E_K}. \tag{22}$$

But as it was shown in [22], this contribution has the magnitude $\sim d_eZ^3\alpha^4/m_p$ and is negligibly small. We disregard it in this work.

For calculating the contribution of the electron EDM (described by the operator H_B) to the atomic EDM, we use the same approach as for studying the e-N interaction. We denote the contribution to the atomic EDM due to H_B as $\mathbf{d}_{\rm at}^B=d_{\rm at}^B(\mathbf{I}/I)$. Using Eq. (1) and replacing $H\to H_B$, after simple transformations we arrive at the following expression:

$$d_{\rm at}^B = d_e\frac{\mu}{3m_pc}\sum_{a,k}\frac{\langle k||r||a\rangle\langle k||H_B^{\rm el}||a\rangle}{\varepsilon_k-\varepsilon_a}. \tag{23}$$

Here, $H_B^{\rm el}$ is the electronic part of the operator H_B. The explicit expression for the ME of the operator $H_B^{\rm el}$ is given in the Appendix.

The results obtained for this contribution to the atomic EDMs is presented in Table V. Comparing our results obtained for Xe and Hg with those of Maartensson-Pendrill and Öster, we see a very good agreement between them. It is seen that the RPA corrections change the results obtained in the DHF approximation for Yb, Hg, and Ra very significantly. For Yb and Ra the DHF and the RPA results differ by a factor of 5. This behavior is quit similar to what we found for $d_{\rm at}^T$, $d_{\rm at}^P$, and $d_{\rm at}^{SM}$ (see Tables II–IV). It is not surprisingly if we take into account the similar nature of all these (P,T)-odd operators. To the best of our knowledge there are no other available data for Yb, Rn, and Ra.

There is also the third-order contribution to atomic EDM proportional d_e [22], which is actually larger than the second-order contribution discussed here. We will consider it in a separate publication.

TABLE VI. The removal energies for both $6s$ electrons for Yb and Hg and both $7s$ electrons for Ra are presented in the first row of each respective atom. Energies of excited states are presented (in cm^{-1}) with respect to the ground state. The results are obtained in the CI and CI+MBPT approximations.

	Config.	Level	CI	CI+MBPT	Experiment
Yb	$6s^2$	$^1S_0$	138795	148707	148712
	$6s6p$	$^3P^o_0$	14368	17562	17288
	$6s6p$	$^3P^o_1$	15029	18251	17992
	$6s6p$	$^3P^o_2$	16537	19995	19710
	$6s6p$	$^1P^o_1$	24215	25715	25068
Hg	$6s^2$	$^1S_0$	213969	235409	235469
	$6s6p$	$^3P^o_0$	30676	37537	37645
	$6s6p$	$^3P^o_1$	32446	39375	39412
	$6s6p$	$^3P^o_2$	36541	44405	44043
	$6s6p$	$^1P^o_1$	48195	54116	54069
Ra	$7s^2$	$^1S_0$	116301	124316	124416
	$7s7p$	$^3P^o_0$	10706	13108	13078
	$7s7p$	$^3P^o_1$	11587	14003	13999
	$7s7p$	$^3P^o_2$	13892	16693	16686
	$7s7p$	$^1P^o_1$	19011	20597	20716

B. CI+MBPT

We use the CI+MBPT approximation for the calculations for Yb, Hg, and Ra atoms, which have two s electrons above closed shells. For the noble gases such as Xe and Rn, the RPA is known to be good for describing their properties (see, e.g., [25]) and there is no need to use the CI+MBPT for them.

We start the discussion of the properties of Yb, Hg, and Ra from the results obtained for the low-lying energies of these atoms. In Table VI we present the energy-level values obtained in the pure CI and the CI+MBPT approximation. As seen from Table VI the removal energies of the two s electrons differ by ~10% from the experimental values at the CI stage. An accounting for the MBPT corrections leads to almost ideal (better than 0.1%) agreement between these quantities. The energies of the excited states (calculated relatively to the ground state) are also noticeably improved at the CI+MBPT stage. The differences between theoretical and experimental values do not exceed 3%. These results indicate the accuracy of wave functions produced at different stages of the CI+MBPT method.

The calculations of the atomic EDM $\mathbf{d}_{\rm at}$ in the CI+MBPT approach are more complicated than in the RPA method. Again we start from Eq. (1) keeping in mind that the summation in this equation is going over many-electron states. Following [26,27] $d_{\rm at}$ can be divided into two parts:

$$d_{\rm at} = d^v_{\rm at} + d^{\rm core}_{\rm at}, \tag{24}$$

where $d^v_{\rm at}$ includes excitations of valence electrons and $d^{\rm core}_{\rm at}$ includes excitations of core electrons and a correction to $d^{\rm core}_{\rm at}$ that appears because of possible excitations of core electrons into the closed valence s shell, which is forbidden by the Pauli principle. Note that the correction restoring the Pauli principle is not small. In certain cases it constitutes ~50% of the total core contribution.

TABLE VII. The valence and core and contributions to $\mathbf{d}^T_{\rm at}$ (in $10^{-20}C_T\langle\boldsymbol{\sigma}_N\rangle|e|$ cm), $\mathbf{d}^P_{\rm at}$ (in $10^{-23}C_P\langle\boldsymbol{\sigma}_N\rangle|e|$ cm), $d^{\rm SM}_{\rm at}$ (in $10^{-17}[S/(|e|{\rm fm}^3)]|e|$ cm), and $d^B_{\rm at}$ (in $10^{-4}d_e$) are presented. The entry "total" means the sum of the valence and core contributions. The accuracy of the values total is estimated at the level of 15–20 %. The results obtained in the RPA method (V^N approximation) are given for comparison. We denote this entry as "RPA."

		$\mathbf{d}^T_{\rm at}$	$\mathbf{d}^P_{\rm at}$	$d^{\rm SM}_{\rm at}$	$d^B_{\rm at}$
^{171}Yb	Val.	−4.24	−14.2	−2.43	6.24
	Core	0.54	1.8	0.31	−0.79
	Total	−3.70	−12.4	−2.12	5.45
	RPA	−3.37	−10.9	−1.95	5.05
^{199}Hg	Val.	−5.71	−20.5	−2.95	12.0
	Core	0.59	2.1	0.32	−1.3
	Total	−5.12	−18.4	−2.63	10.7
	RPA	−5.89	−20.7	−2.99	12.3
^{225}Ra	Val.	−20.6	−75.0	−10.25	−65.2
	Core	3.0	10.8	1.41	9.5
	Total	−17.6	−64.2	−8.84	−55.7
	RPA	−16.7	−61.0	−8.27	−53.3

With the wave functions obtained from Eq. (13), the valence part $d^v_{\rm at}$ is computed with the Sternheimer [28] or the Dalgarno-Lewis [29] method implemented in the CI+MBPT+RPA framework. Given the wave function $|0\rangle$ and its energy E_0, we find an intermediate-state wave function $\Phi_{\rm in}$ from the inhomogeneous equation

$$|\Phi_{\rm in}\rangle = \frac{1}{H_{\rm eff} - E_0}\sum_K |K\rangle\langle K|(r_z)_{\rm eff}|0\rangle = \frac{1}{H_{\rm eff} - E_0}(r_z)_{\rm eff}|0\rangle. \tag{25}$$

Given $\Phi_{\rm in}$ we can compute $d^v_{\rm at}$ as

$$d^v_{\rm at} = 2\langle 0|(r_z)_{\rm eff}|\Phi_{\rm in}\rangle. \tag{26}$$

An additional contribution $d^{\rm core}_{\rm at}$ coming from particle-hole excitations of the core is incorporated in the frame of the RPA approach discussed above.

In Table VII we list the results obtained for the valence and the core contributions to the EDM of the atoms. For completeness and comparison we also present the values obtained in the RPA calculations. As seen from the table the largest differences between the results obtained in the RPA and the CI+MBPT methods occur for Hg. But even in this case these differences do not exceed 15%. Taking into account the similar nature of the (P,T)-odd operators considered in this work, we can expect that the relative difference between the results found in both approximations would be

approximately the same for all operators for a given atom. As we see from Table VII, this condition is fulfilled. This is a good consistency test of the calculations.

In this way we arrive at the conclusion that the electron correlations do not affect the final results too much. Based on this observation we estimate the accuracy of the values "total" listed in Table VII is at the level of 15–20 %. Similar accuracy is expected for Xe and Rn atoms.

V. SHELL MODEL OF THE NUCLEUS

In certain cases it is possible to impose additionally the constraints on the strength of the couplings for protons and neutrons. Below we will obtain such constraints for a simple case of the spherical shell model of a nucleus. This model describes well the nuclei of ^{129}Xe and ^{199}Hg. The former contains a neutron in the $s_{1/2}$ state and the latter contains a neutron in the $p_{1/2}$ state above the closed shells. Although the nuclei in the atoms have a valence neutron, it is possible to deduce also the proton contribution to the total nuclear spin using the information on the nuclear magnetic moments. If we assume that the magnetic moment of the nucleus is composed entirely from the spin magnetic moment of the valence neutron and the spin magnetism of polarized nuclear core, then

$$\mu = \mu_n \langle \sigma_z^{(n)} \rangle + \mu_p \langle \sigma_z^{(p)} \rangle,$$

$$\langle \sigma_z^{(n)} \rangle + \langle \sigma_z^{(p)} \rangle = \langle \sigma_z^{(0)} \rangle. \tag{27}$$

Here, $\mu_p \approx 2.793$ and $\mu_n \approx -1.913$ are the magnetic moments of the proton and the neutron expressed in nuclear magnetons. Numerical estimates show that the main contribution to the nuclear magnetic moments μ of ^{129}Xe and ^{199}Hg comes from the neutron and proton spin contributions. This is due to that the neutron orbital contribution is zero and the proton orbital contribution (for the orbitals with a low orbital momentum) is small in comparison with its spin contribution. Neglecting the spin-orbit interaction leads to conservation of the total spin, which is equal to the average spin of the neutron above the unpolarized core $\langle \sigma_z^{(0)} \rangle$. Taking into account that, in the spherical shell model the nuclear spin $\mathbf{I}$ is determined by the total momentum of the unpaired nucleon, we can write

$$\langle \boldsymbol{\sigma}_N \rangle = \langle \sigma_z^{(0)} \rangle \mathbf{I}/I, \tag{28}$$

with

$$\langle \sigma_z^{(0)} \rangle = \begin{cases} 1, & I = l_I + 1/2 \\ -I/(I+1), & I = l_I - 1/2, \end{cases}$$

where l_I is the orbital quantum number of the valence nucleon. Using Eq. (28), we determine $\langle \sigma_z^{(n)} \rangle$ and $\langle \sigma_z^{(p)} \rangle$ for observationally relevant cases of ^{129}Xe and ^{129}Hg as shown in Table VIII.

TABLE VIII. Composition of the nuclear spin.

Nucleus	Neutron state	$\langle \sigma_z^{(0)} \rangle$	$\langle \sigma_z^{(n)} \rangle$	$\langle \sigma_z^{(p)} \rangle$
^{129}Xe	$s_{1/2}$	1	0.76	0.24
^{199}Hg	$p_{1/2}$	−1/3	−0.31	−0.03

TABLE IX. The recommended values of the contributions to atomic EDM.

	^{129}Xe	^{171}Yb	^{199}Hg	^{211}Rn	^{225}Ra
$\mathbf{d}_{\rm at}^T$ $(10^{-20} C_T \langle \boldsymbol{\sigma}_N \rangle / \lvert e \rvert$ cm)	0.57	−3.7	−5.1	5.6	−18
$\mathbf{d}_{\rm at}^P$ $(10^{-23} C_P \langle \boldsymbol{\sigma}_N \rangle / \lvert e \rvert$ cm)	1.6	−12	−18	21	−64
$d_{\rm at}^{\rm SM}$ $\{10^{-17}[S/(\lvert e \rvert {\rm fm}^3)]\lvert e \rvert$ cm$\}$	0.38	−2.1	−2.6	3.3	−8.8
$d_{\rm at}^B$ $(10^{-4} d_e)$	1.0	5.5	11	−13	−56

As seen from Table VIII the contribution of the proton spin into the total nuclear spin of ^{129}Xe is as high as 30%, and therefore the proton couplings C_T^p and C_P^p [see the equations below Eq. (5)] are also limited in the experiments searching for the EDM. For ^{199}Hg the limit on the proton couplings is ten times weaker than the limit on the neutron couplings.

Note that the simple shell model of a nucleus considered above is hardly applicable to the nuclei of ^{171}Yb, ^{211}Rn, and ^{225}Ra. The problem is that the nucleus of ^{171}Yb is quadrupole deformed and the nuclei of ^{211}Rn and ^{225}Ra are octupole deformed. For this reason more sophisticated nuclear models are required for a proper description of these nuclei.

VI. CONCLUSION

In conclusion, we have carried out calculations of different contributions to the EDMs of ^{129}Xe, ^{171}Yb, ^{199}Hg, ^{211}Rn, and ^{225}Ra by the RPA and the CI+MBPT methods. Two of these contributions are due to tensor-pseudotensor and scalar-pseudoscalar e-N (P,T)-odd interactions, and two more contributions are caused by the nuclear Schiff moment and the electron EDM. The recommended values for noble gases Xn and Ra are based on the results obtained in the RPA calculations while for Yb, Hg, and Ra they based on the calculations carried out in the frame of CI+MBPT+RPA approach. These numbers are gathered in Table IX.

Finally, using the results obtained in this work for atomic mercury and recently obtained upper bound $|d_{\rm at}(^{199}{\rm Hg})| < 3.1 \times 10^{-29} |e|$ cm [4], we are able to find limits on CP-violating parameters C_T, C_P, and S. We do not put a constraint on the coupling constant d_e because the contribution of the electric dipole electron moment to the atomic EDM in the third order of the perturbation theory is known to be ~10 times larger than its contribution in the second

TABLE X. Limits on CP violating parameters C_T, C_P, and S based on an experimental limit for $|d_{\rm at}(^{199}{\rm Hg})| < 3.1 \times 10^{-29} |e|$ cm [4].

Parameter	Limit
C_T	1.9×10^{-9}
C_P	5.2×10^{-7}
S $(\lvert e \rvert {\rm fm}^3)$	1.2×10^{-12}

order [22]. It means that a dominating contribution from the electron EDM still has to be considered. This is a subject of another work. Applying again the spherical shell model to ^{199}Hg and, respectively, having $\langle \boldsymbol{\sigma}_N \rangle = -\frac{1}{3}(\mathbf{I}/I)$, we arrive at the numbers listed in Table X.

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APPENDIX

To calculate the MEs of the (P,T) operators we define the one-electron wave function $|a\rangle \equiv \psi_a(\mathbf{r})$ as follows:

$$\psi_a(\mathbf{r}) = \begin{pmatrix} f_a(r)\Omega_{j_a l_a m_a}(\mathbf{n}) \\ i g_a(r)\Omega_{j_a \tilde{l}_a m_a}(\mathbf{n}) \end{pmatrix}, \tag{A1}$$

where $\tilde{l}_a = 2j_a - l_a$.

The MEs of the (P,T)-odd operators characterizing the tensor-pseudotensor and the pseudoscalar-scalar interactions are given by the following expressions:

$$\begin{aligned} &\langle n_b\kappa_b || i\gamma\rho(r) || n_a\kappa_a \rangle \\ &= \langle \kappa_b || C_1 || \kappa_a \rangle \int_0^\infty \{ f_b g_a (\kappa_a - \kappa_b + 1) \\ &\quad + f_a g_b (\kappa_b - \kappa_a + 1) \} \rho(r) r^2 dr, \end{aligned} \tag{A2}$$

$$\begin{aligned} &\langle n_b\kappa_b || n(d\rho/dr)\gamma_0 || n_a\kappa_a \rangle \\ &= \langle \kappa_b || C_1 || \kappa_a \rangle \int_0^\infty (g_b g_a - f_b f_a) r^2 \frac{d\rho}{dr} dr. \end{aligned} \tag{A3}$$

If the nuclear density distribution is given by Eq. (5) (see the main text) Eq. (A3) is further simplified leading to

$$\langle n_b\kappa_b || n(d\rho/dr)\gamma_0 || n_a\kappa_a \rangle = \langle \kappa_b || C_1 || \kappa_a \rangle \frac{3}{4\pi} \left(\frac{g_b g_a - f_b f_a}{r} \right)_{r=R}, \tag{A4}$$

where spherical harmonics C_{lm} are defined as

$$C_{lm}(\mathbf{n}) = \sqrt{\frac{4\pi}{2l+1}} Y_{lm}(\mathbf{n}), \tag{A5}$$

$\kappa = (l-j)(2j+1)$, and the reduced ME $\langle \kappa_b || C_1 || \kappa_a \rangle$ is given by

$$\begin{aligned} \langle \kappa_b || C_1 || \kappa_a \rangle &= \xi(l_b + l_a + 1)(-1)^{j_b + 1/2} \sqrt{(2j_a+1)(2j_b+1)} \\ &\quad \times \begin{pmatrix} j_b & j_a & 1 \\ -1/2 & 1/2 & 0 \end{pmatrix}, \end{aligned} \tag{A6}$$

where

$$\xi(x) = \begin{cases} 1, & \text{if } x \text{ is even} \\ 0, & \text{if } x \text{ is odd.} \end{cases}$$

The reduced ME characterizing the nuclear Schiff moment is given by

$$\langle n_b\kappa_b || r\rho(r) || n_a\kappa_a \rangle = \langle \kappa_b || C_1 || \kappa_a \rangle \int_0^\infty \{ g_b g_a + f_b f_a \} r^3 \rho(r) dr. \tag{A7}$$

The reduced ME of the operator $H_B^{\rm el}$ can be represented as

$$\begin{aligned} &\langle n_b\kappa_b || H_B^{\rm el} || n_a\kappa_a \rangle \\ &= -\langle \kappa_b || C_1 || \kappa_a \rangle \left[\int_R^\infty \{ f_b g_a (\kappa_b - \kappa_a + 2) \right. \\ &\quad + f_a g_b (\kappa_a - \kappa_b + 2) \} \frac{dr}{r} - 2 \int_0^R \{ f_b g_a (\kappa_b - \kappa_a - 1) \\ &\quad \left. + f_a g_b (\kappa_a - \kappa_b - 1) \} \frac{r^2 dr}{R^3} \right]. \end{aligned} \tag{A8}$$

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